The Impact of Tropospheric Rocket Exhaust on Stratospheric Ozone

Prepared for:

Environmental Management Division
Space and Missile Systems Center
El Segundo, California

Prepared by:

TRW Space and Electronics Group

in coordination with:

Michael J. Prather, Ph.D.
Professor, Department of Geosciences
University of California, Irvine

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Approved by:

John J. Lamb, Ph.D.
Program Manager

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ABSTRACT

The relative importance of direct stratospheric injections of chlorine from the Solid Rocket Motors on the Space Shuttle and other spacecraft such as the Titan IV have been assessed previously. The possibility that the tropospheric exhaust (2/3 of the total for the Shuttle SRM) would eventually enter the stratosphere and contribute to ozone depletion has not been evaluated. A three-dimensional chemical transport model is used to simulate the fate of the tropospheric component of the Shuttle exhaust: its dispersion, wet removal in convective events, and possible entrainment into the stratosphere. For any reasonable value for the efficiency of removal in deep, wet, cumulus convection, more than 99.5% of the soluble chlorine injected below 200 mbar is removed in the first three months. The fraction entering the stratosphere is less than 0.2% in both summer and winter seasons. Additional evidence from microphysical models and from analyses of the chlorine budget of the stratosphere argues that soluble chlorine from the troposphere cannot contribute significantly to stratospheric chlorine. In summary, only direct stratospheric injection of chlorine (i.e., fuel burned in the stratosphere) is potentially important; the tropospheric emissions from perchlorate-fueled SRM (e.g., Space Shuttle, Titan IV) are unlikely to impact stratospheric ozone.

1. Introduction

Launches of the Space Shuttle and other perchlorate-fueled launch vehicles such as the Titan IV emit soluble chlorine compounds along the launch path through the atmosphere while the solid rocket boosters are burning. The effects of emissions from the Titan IV and the Space Shuttle released directly into the stratosphere are well-known and have been detailed elsewhere (WMO, 1992; Prather et al., 1990a; AIAA, 1991, Ko et al., 1994). Here, we focus on how the tropospheric emissions may be transported into the lower stratosphere before being removed by washout through wet convective processes. In particular, we seek to answer the following questions:

- How rapidly is the original material removed by wet convective processes?
- Where is it removed relative to the launch path?
- How much is transported into the stratosphere, where, and by what means?
- How sensitive are these results to the rate of removal specified?
- Are these results sensitive to seasonal differences?

The major effort here is in the use of a three-dimensional atmospheric model to follow the solid rocket motor exhaust in the lower atmosphere. In our conclusions, we also review other evidence showing that soluble chlorine (as HCl) do not enter the stratosphere in significant quantities.

2. Atmospheric Model

2.1 Background

The analysis which follows is based on the use of a three-dimensional chemical tracer model (CTM) which uses meteorological data from a general circulation model as input. The input data is derived from the Goddard Institute for Space Sciences general circulation model (Hansen et al., 1983) and the CTM, in its tropospheric 9-layer version, has been used in a number of studies to simulate distributions and lifetimes of trace species, including chlorofluorocarbons (Prather et al., 1987), methyl chloroform (Spivakovsky et al., 1990), and ozone and its precursors (Jacob et al., 1993). The version of the CTM used in this analysis has 21 layers and has been used in studies of carbon dioxide, ozone and other species from meteorites (Rind et al., 1990; Prather and Rodriguez, 1988; Prather et al., 1990b; Hall and Prather, 1993).

There has been some investigation into the usefulness of the CTM in representing the transport and scavenging of soluble species. Balkanski et al. (1993) considered the scavenging of ^{210}Pb, a decay product of ^{222}Rn emitted from soils, using a three-dimensional tracer model similar to the one used here. Scavenging was incorporated directly into the mass transport operator, coupling the removal and transport processes, and the "scavenging efficiency", the fraction

of aerosol lost to deposition before dispersing at the top of the convective column, was set at 50% for shallow (less than 2600 m altitude) convection, and 100% for deep convection. (Balkanski, et al, 1993) It was found that 74% of the global ^{210}Pb sink was represented by scavenging in convective updrafts, with 12% occurring in large-scale precipitation events, and the remaining 14% due to dry deposition.

For comparison, a similar analysis by Feichter et al. (1991) found 61% removal by large-scale precipitation, 26% removal by convective events, and 13% removal by dry deposition. However, Feichter et al. (1991) used a simpler first-order rainout scheme where surface concentrations and surface deposition fluxes were found to disagree sharply with observed values. The improvements made by Balkanski et al. (1993) addressed these shortcomings.

2.2 Documentation

The CTM solves the mass conservation equations for HCl, the soluble compound used in this study, on a global three-dimensional grid. This grid matches the grid used in the development of the meteorological input data, and has 7.83 degree latitude by 10 degree longitude resolution in the horizontal, for a total of 36x24 grid points. The CTM has 21 layers in the vertical extending from the surface to the top of the atmosphere, at 0.022 mb, using sigma coordinates in the nine layers to 100 mb, and pressure coordinates above 100 mb. A one-year archive of GCM data serves as the primary input to the model, and for multi-year runs, the model recycles the one year of archived GCM data. The model uses eight-hour averages of horizontal velocities, determines vertical velocity from convergence calculations, and has convective patterns for each grid archived on a monthly basis by the GCM.

The mass transport computation is that of Prather et al. (1986; 1987). The mass fluxes due to convection are determined by the same scheme as is used in the GCM itself (Hansen et al., 1983): when a column of atmosphere is unstable with respect to wet convection, half of the air mass in the lowest layer is transported (condensing and precipitating along the path) to the layer at which it is neutrally buoyant without any entrainment. This is followed by subsidence from the top through the middle layers as needed to conserve mass. When a column of atmosphere is unstable with respect to dry convection only (i.e. large-scale instability associated with fronts), the entire mass of air within the column is mixed uniformly.

Loss of soluble species due to wet convection is crudely parameterized through the use of a "wet removal efficiency". It represents the fraction of tracer removed by deposition on its path through the convective column. This value is applied in the convective mass transport operator as a sink term whenever wet convection occurs, as wet convective events in the GISS GCM are always associated with precipitation. This follows from the idea that vertical motion and

the associated condensation of water is an effective removal mechanism of soluble species (Tabazadeh and Turco, 1993). This mechanism may not result in removal if the precipitation evaporates at some lower level. Large scale precipitation can also remove soluble species from the layer in which the cloud forms, as well as through the pick-up of soluble compounds by the falling hydrometeors. This form of precipitation is diagnosed by the GCM but has not been used by the CTM for tracer removal. Additional loss of a reactive or soluble species from the atmosphere occurs with contact of the surface (i.e. dry deposition). In general, these species, such as HCl, are not re-volatilized. Previous numerical experiments have found that the dominant loss processes are convective (particularly in the tropics and mid-latitudes during summer), and argue that large-scale precipitation losses are minimal (Balkanski et al., 1993). Most importantly, the calculations presented here can be considered as an upper bound on the stratosphere source in the absence of additional losses due to dry deposition and large-scale non-convective precipitation.

2.3 Numerical Simulation

2.3.1 Emissions

Table 1 gives the emission data, in metric tons of chlorine (as HCl) for a single launch of the Space Shuttle. We have chosen the Shuttle Solid Rocket Motors as an example here; results would apply equally to the Titan IV SRM, for example. From this data, mass inputs for each of the CTM layers were computed. The emissions from a single launch were represented as an injection in the form of a single column "puff" of chlorine as HCl, at the start of each model run, positioned in the 8 x 10 grid box corresponding to Cape Canaveral, Florida (28.61 N, 80.68 W). Background concentrations were initialized near zero in all other grid boxes of the model domain.

2.3.2 Diagnostics

To assist in answering the questions detailed above, two new model diagnostics were developed. The first used existing CTM inputs to define a diagnostic "tropopause" boundary to monitor exchange between the model troposphere and the model stratosphere. The experiments were conducted with this boundary defined at the top of model layer 8 (206 mb) for latitudes poleward of 24 degrees on either side of the equator, and as the top of layer 9 (100 mb) for the tropical regions between 24 degrees north and south of the equator. This boundary was developed to better represent the actual mean positions of the tropical and mid-latitude tropopause, and to assist in diagnosing stratosphere-troposphere exchange resulting from transports across the mid-latitude break in the tropopause at the level of the jet stream. The definition of such a boundary is indeed arbitrary and is presented in Figure 1. We can use flux diagnostics already present in the CTM on this "boundary level" and determine the vertical and horizontal exchange across this arbitrary boundary. However, the diagnostic

"tropopause" as defined here is a boundary which does not change seasonally and does not respond to synoptic variations in the height of the tropopause. Elements of the eighth CTM layer, a layer we specify as "tropospheric", may actually contain elements of atmosphere which are "stratospheric" by nature of the CTM. We would expect longer "stratospheric" residence times and less removal by wet convection from such regions. As we shall see, results from experiments conducted on chlorine (as HCl) emitted in this eighth layer exhibit more of a "stratospheric" character than those experiments conducted on chlorine (as HCl) emitted at lower altitudes in the CTM. The eighth layer should be considered as a "transition" region between the model troposphere and stratosphere, rather than a layer which is entirely contained in the model troposphere.

The second diagnostic allows determination of the changes in tracer mixing ratio within a specific volume (i.e. a "box" of atmosphere), the boundaries of which are specified. This diagnostic also notes which process (advection, diffusion, or wet removal) alters the mixing ratio within the box. When combined with the "tropopause" diagnostic as a box in the elevated tropical troposphere layer (layer 9 between 24 N and 24 S), this allows determination of the flux of tracer through the vertical boundaries of the model "tropopause", crudely simulating exchange occurring in tropopause discontinuities at the subtropical jet (Figure 1). The numerical accuracy of the "box" approach was verified by specifying the boundaries of the box as the boundaries of the model grid, and observing no loss of tracer by advective or diffusive processes.

These two diagnostics allow us to answer questions with regards to transport into the stratosphere from the tropospheric emissions, for we can diagnose both vertical and horizontal transports which occur across our diagnostic tropopause. Spatial patterns of rainout and tracer budgets for the entire domain are part of the existing diagnostics in the CTM.

2.3.3. Two-Year Runs

The first set of experiments were conducted for two simulated model-years following a launch to determine the sensitivity of the results to changes in the wet removal efficiency parameter. The model was begun on July 1. The entire column of chlorine emissions were used in this first case, as given in Table 1. The model simulations were made for ten different wet removal efficiencies to determine sensitivity to the rate chosen. Diagnostics consisted of one chart which detailed the partitioning of the tracer mass between the model troposphere and stratosphere layers, and a time series taken at a grid point downstream of the launch column and slightly above the top of the model troposphere (65 W, 43 N, and 73 mb). Total tracer budgets were also monitored to determine instantaneous lifetimes at the end of each month, by comparing the change in the total chlorine present in the model domain to the entire amount.

2.3.4. Three-Month Runs

The second set of experiments were conducted over a shorter period of time, based on the analysis of the first set of two-year experiments. Since most (greater than 95%) of the tropospheric chlorine (as HCl) was removed rapidly in the two-year simulations, these calculations focus on the removal processes for tracer emitted into each of the tropospheric model layers (1-8, surface to 200 mb) over a much smaller time period with more intensive diagnostics. These diagnostics consisted of the detailed partitioning of the tracer mass in the model domain (as described above), a latitude-longitude plot of accumulated removal through wet convection, a latitude-longitude plot of accumulated net transport to or from the stratosphere, determined across the "tropopause" diagnostic defined earlier. Using these diagnostics, the fate of all of the tropospheric chlorine emitted by the Solid Rocket Motors during the first three months after launch was determined.

The wet removal efficiency for these three-month simulations was fixed at 75% based on the optimization used by Balkanski et al., (1993) who used 50% for shallow convection (below layer 3) and 100% for deep convection. Results from the two-year runs showed that for a large range of removal efficiencies, the evolution of total mass of HCl is insensitive to changes in the removal rate specified. Sixteen total experiments were conducted, one for each layer of the lower atmosphere (layers 1-8) for both a January 1 release and a July 1 release. These were chosen so as to investigate the seasonal effects on the distribution and removal, and also to find how emissions in each of the individual layers behaved.

3. Results

3.1. Two-Year Runs

The results of the two-year simulations demonstrate the insensitivity of results to the removal rate specified. Ten experiments were conducted, each with a different removal rate (0, 2, 4, 6, 8, 10, 35, 50, 75, and 100% wet removal efficiency).

For a selected grid point downwind of the launch column (65W, 43N, 73 mb), a time series of the difference in mixing ratios between a 0% removal rate and a 100% removal rate is shown in Figure 2. The mean mixing ratio throughout these experiments was 77 ppt for the 0% case, and 62 ppt for the 100% case. The difference between these two simulations reaches 20 ppt after 20 months. These results represent the extreme case: tropospheric emissions could contribute at most 30% to the stratospheric HCl levels resulting from perchlorate-fueled launches in the unlikely event that none of the tropospheric emissions were removed.

Time series plots of the total amounts of tracer remaining in the model domain are given in Figures 3a and 3b. For removal rates above 35%, the calculations yield the same overall tracer mass, to within 5%, after the first two or three months of the calculation. Beyond six months, all calculations except those corresponding to extremely insoluble (i.e. wet removal efficiencies of less than 4%) tracers converge to the same overall mass amounts. Thus, for the moderately soluble forms of chlorine under study here, it is clear that the results of our experiments are insensitive to the removal rate chosen.

Additionally, plots of the instantaneous "lifetime" (total burden divided by total sink per unit time) confirm that, for removal rates greater than or equal to 35%, the instantaneous lifetime does not change appreciably (Figures 4a and 4b). As expected, plots of this quantity reflect the relative ease with which a soluble compound is removed during the first year of the experiment, and the difficulty of removal in the second year reflected as an increased "lifetime" resulting from residence of a small remaining fraction in the stratosphere.

In summary, the results of the two-year runs demonstrate that, for a moderately soluble tracer, both local and global results from the CTM are relatively insensitive to the choice of wet removal efficiency, provided its value is at least 35%.

3.2 Three-month Runs

These short-term simulations allowed for more detailed diagnosis of the fate of chlorine deposited in each tropospheric model layer. These diagnostics examined the spatial distribution of exchange across the arbitrarily specified "tropopause", and the spatial distribution of removal by wet deposition.

3.2.1 Global Budgets

The fates of the emissions for January and July are detailed in Table 2, which shows for each layer the mass amounts transported into the stratosphere in the vertical, the amount transported into the stratosphere through tropopause discontinuities meridionally ("across the jets"), the amount remaining in the stratosphere at the end of the three-month period, and the amount removed from the domain through wet deposition.

In all cases studied here, less than 1% of the emitted chlorine (as HCl) makes it into the model stratosphere. This fraction decreases to less than 0.2% if only the lowest seven layers are considered (i.e. emissions below 350 mb). Dominant processes which transport chlorine (as HCl) to the stratosphere are horizontal through the subtropical break in the tropopause in January, and vertical due to deep convection in July. However, it is not clear that such transport is truly into the stratosphere since our diagnostic definition is not exact.

The most complete removal of chlorine (as HCl) occurs in January, particularly of those fractions emitted in the lowest four layers. The total amount of chlorine (as HCl) transported to the stratosphere from emissions in the lowest seven layers of the CTM troposphere was calculated to be 8.55×10^4 kg for January, and 11.844×10^4 kg for July. These amounts represent an additional 0.10% (January) or 0.14% (July) increase to the emissions placed directly into the stratosphere by the solid rocket boosters.

The evolution of the total mass of chlorine remaining in the model domain is detailed for some of the three month experiments in the Figure 5. The wet removal efficiency for these simulations is fixed at 75%. For all of the January launches, fractions released in the lowest layers (1-3) were reduced to less than 1% of their initial mass by wet deposition within three weeks after launch. This time period for reduction to 1% of the original mass steadily increases with height of the layer. In particular, releases in the uppermost layers of the CTM troposphere (up to layer 7) take somewhat longer, on the order of five to six weeks, to be removed to the 1% level. For July launches, the time periods involved are somewhat longer, on the order of five to six weeks for removal to the 1% level. Unlike the January launches, however, the time involved remains fairly constant with altitude of the initial release, through layer 7. Emissions released in the eighth CTM layer (200-350 mbar) are removed more slowly during the three-month period, with final fractions of remaining mass being about 2-3% for both January and July. The change in slope of the curves of Figure 5 indicate an increasing residence time for the remaining chlorine (i.e. a straight line on this semi-log plot corresponds to an exponential decay). The time scale for removal of the remaining mass approaches two years, and thus only this small fraction (less than 0.2% for emissions below 350 mb) appears to have entered the stratosphere.

3.2.2 Wet Removal

Figures 6 through 9 show the patterns of wet removal and stratosphere-troposphere exchange for the emissions at the different model levels.

For the January launch, emissions in the lowest five layers (below 660 mb) are rained out along a northeast to southwest line centered on the launch site, with most significant amounts being deposited within the grid box where launch occurred. At middle levels (5 and 6 for the CTM, 350-660 mb) the chlorine material is transported further before rainout occurs, as evidenced by the stretching of the accumulated wet deposition contours in Figure 6. The localized area of large removal near the launch site is distinct through layer 7 of the CTM, although substantially more chlorine reaches the northern tropics in higher layers. Emission fractions in layer 8 (200-350 mb) are spread throughout the model domain before removal. The southernmost extent of removal varies from about 20 degrees South Latitude in layer 1 to the entire Southern Hemisphere in layer 8 (just below the arbitrary tropopause).

July removal patterns are similar to those for January, with some exceptions, as shown in Figure 7. In all layers, the intense area of deposition near the launch site is less distinct, and there is more wet removal taking place downstream directly to the east of the launch point. Maximum removal over the launch site does occur in CTM layer 4, as it did for January, and the magnitudes are roughly equal. However, removal of the chlorine from layer 1 occurs as far south as 50 degrees South Latitude, further south than for the January launch. Noticeable removal occurs throughout the Southern Hemisphere for emissions released in layer 8 (just below the arbitrary tropopause).

3.2.3. Exchange between Stratosphere and Troposphere

Due to the arbitrary specification of the "tropopause" used in this study, assessment of the exchange between the troposphere and the stratosphere should be viewed with caution. A more detailed study of these exchanges would include a seasonally varying "tropopause" (e.g. one based on potential temperature) to reflect its true transient positions. At best, these results should be considered qualitative.

For the January launch, exchange between the troposphere and the stratosphere is minimal for the emission in layers 1-7, and negligible for the lowest three layers. The exchange between the troposphere and stratosphere is much greater for layer 8, nearest the boundary located at 206 mb. Maps of vertical exchange between the two regions are shown in Figure 8. Nearly all of the significant exchange takes place along the latitude band of the original release, with pockets of exchange into the stratosphere located over the launch site, and near 100 degrees West Longitude. Very little exchange occurs in the tropics or south of the equator.

Greater apparent transport of chlorine into the stratosphere takes place in for the July launch, with largest transports downstream of the launch site in the same latitude band. This may be partially as a result of the diagnostic boundary chosen, as the actual atmospheric tropopause may be much higher in altitude than the boundary chosen here. The maps of exchange for July are shown in Figure 9. Comparing with the results from the January launches, however, the lower layer fractions contribute to more of the total exchange in July than they do in January. This may be as a result of the GCM meteorology, with deeper convection present in the summer months (July) over the northern mid-latitudes, rather than the stratiform precipitation which is present throughout the winter months (e.g. the January launch).

4. Conclusion

It is highly unlikely that soluble chlorine (as HCl) emitted into the troposphere by perchlorate-fueled solid rocket boosters (e.g. Titan IV, Space

Shuttle) will enter into the stratosphere in significant quantities. Numerical simulations using a chemical tracer model using a most likely parameterization of wet removal in convective events (75% efficiency) show that less than 0.5% of the original tropospheric emissions remain in the atmosphere after three months, and less than 0.2% of these original tropospheric emissions are transported across the diagnostic "tropopause" into the lower stratosphere. Greater than 99.5% of the original emissions are removed by washout, most of which occurs in the first six weeks after launch. Removal occurs primarily nearest the point of launch, with some removal taking place downstream from the launch site. Only emissions above 600 mb transport a noticeable fraction into the tropics and these are also removed by wet convection before entering the stratosphere.

The ability of deep, wet convection to remove soluble species before they enter the stratosphere is supported by these model simulations. Even for small efficiencies of convective removal (as low as 6% removal per event from the convective plume), the tropospheric burden is rapidly reduced, and the amount entering the stratosphere is extremely small, less than 0.2% below 350 mb. This fraction is certainly not known to better than a fraction of two, but the upper limit appears robust.

These conclusions are strengthened by other independent approaches. Detailed microphysical models (e.g. Tabazadeh and Turco, 1993) show that soluble chlorine is efficiently removed in ascending volcanic plumes and does not enter the stratosphere in significant quantities. In spite of the presence of tropospheric HCl (about a part per billion), the stratospheric chlorine budget can be balanced by the chlorine entering the stratosphere as organochlorines (e.g. chlorofluorocarbons) (Zander, et al, 1992).Thus, the HCl present in the lower troposphere must be efficiently removed by wet convection before the air is injected into the stratosphere. Since moist air in the lower stratosphere (~ 1% water vapor) must be dehydrated to a few parts per million before it enters the stratosphere, it is difficult to envisage a process that would not remove an equally large fraction of HCl present in solution with water vapor.

In summary, without significantly increasing stratospheric chlorine levels, it is unlikely that the tropospheric emissions of perchlorate-fueled solid rocket boosters could impact stratospheric ozone.

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6. Tables and Figures

Table 1: Cl emissions into 5-km bins from a single Space Shuttle Launch

Table 2: Fate of single-layer emissions of Shuttle Cl, 3 months after launch. For the stratospheric injection data, positive values denote transport into the stratosphere.

Figure 1: Definition of "tropopause" boundary as used in CTM experiments.

Figure 2: Time series of difference in tracer concentrations at 65 W, 43 N, 73 mb between two runs with different "removal efficiencies" (0% wet removal and 100% wet removal).

Figure 3a: Time series of chlorine (as HCl) mass (kg) with time (total hours) for two-year experiments with "removal efficiencies" between 0% and 10%.

Figure 3b: As in Fig. 3a, for the two-year experiments with "removal efficiencies" above 10%.

Figure 4a: Variation of the "instantaneous lifetime" (burden/loss rate) for the two-year experiments with "removal efficiencies" of 2%, 10%, and 35%.

Figure 4b: As in Fig. 4a, for the two-year experiments with "removal efficiencies" of 50%, 75%, and 100%.

Figure 5a: Time series of chlorine mass (in kg HCl) for the single-layer emissions in layer 1. Releases in January and July are plotted on the same axes. Each point represents roughly one model week.

Figure 5b: As in Fig. 5a, for emissions in layer 4.

Figure 5c: As in Fig. 5b, for emissions in layer 7.

Figure 6: Spatial pattern of surface wet deposition (in kg HCl/grid area) for the single-layer emissions in layers 1, 4, and 7 for the three-month period after a January launch.

Figure 7: As in Fig. 6, for the three-month period following a July launch.

Figure 8: Spatial pattern of vertical exchange (in kg HCl/grid area) across the arbitrary "tropopause" defined by Figure 1, for the single-layer emissions in layer 1, 4, and 7 for a January launch.

Figure 9: As in Fig. 8, for the three month period following a July launch.

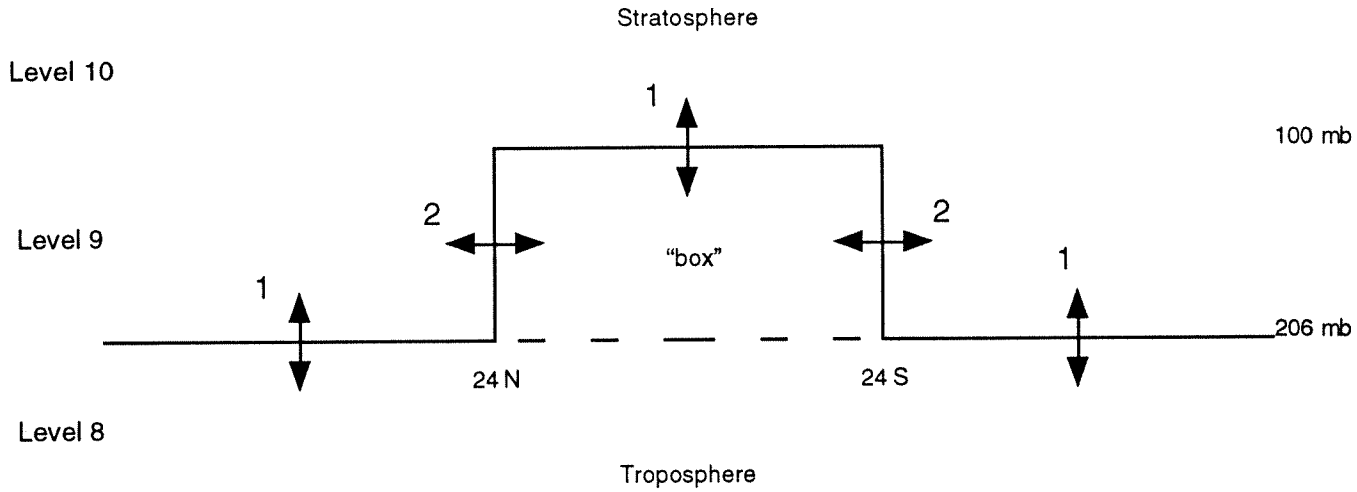
Table 1

Altitude (km)	Cl -Emissions (ton/5 km bin)
0-5	65
5-10	40
10-15	28
15-20	19.6
20-25	15.1
25-30	12.2
30-35	9.7
35-40	7.7
40-45	2.9
45-50	0.5
sum	200.7

Table 2

Launch	Injection Layer	Total Input Mass (kg)	Remaining in Atmosphere (kg)	Pct Removed	Stratosphere Mass (kg)	Stratospheric Injection-Vertical (kg)	Stratospheric Injection-Horizontal (kg)
January 1	1	2.716e+6	6.911e+2	99.975%	3.017e+2	-1.156e+1	3.133e+2
	2	3.649e+6	6.176e+2	99.983%	2.789e+2	-4.678e+1	3.257e+2
	3	5.502e+6	1.395e+3	99.975%	6.140e+2	-4.565e+1	6.596e+2
	4	1.128e+7	5.839e+3	99.948%	2.590e+3	-8.647e+2	3.454e+3
	5	1.929e+7	3.370e+4	99.825%	1.486e+4	1.334e+3	1.353e+4
	6	2.598e+7	6.773e+4	99.739%	2.876e+4	-7.052e+3	3.581e+4
	7	2.243e+7	8.593e+4	99.617%	3.810e+4	-2.632e+4	6.442e+4
	8	2.496e+7	5.089e+5	97.961%	2.290e+5	1.491e+6	-1.262e+6
July 1	1	2.671e+6	8.345e+3	99.688%	4.320e+3	9.048e+3	-4.728e+3
	2	3.589e+6	1.085e+4	99.698%	5.592e+3	1.269e+4	-7.098e+3
	3	5.413e+6	1.671e+4	99.691%	8.651e+3	1.789e+4	-9.239e+3
	4	1.109e+7	3.338e+4	99.699%	1.731e+4	3.283e+4	-1.552e+4
	5	1.898e+7	5.016e+4	99.736%	2.604e+4	3.025e+4	-4.208e+3
	6	2.556e+7	5.726e+4	99.776%	2.839e+4	1.486e+4	1.353e+4
	7	2.207e+7	6.602e+4	99.701%	3.051e+4	1.532e+4	1.519e+4
	8	2.455e+7	4.481e+5	98.175%	2.256e+5	5.954e+5	-3.698e+5

Figure 1:Stratosphere-Troposphere Exchange in the CTM Model



1 - Vertical flux by direct transport

2 - Horizontal transport across subtropical discontinuities in tropopause
(sum of meridional advective and diffusive fluxes out of "box")

Figure 2: Difference in Mixing Ratios with "Removal Effieciency" (0% and 100% experiments)

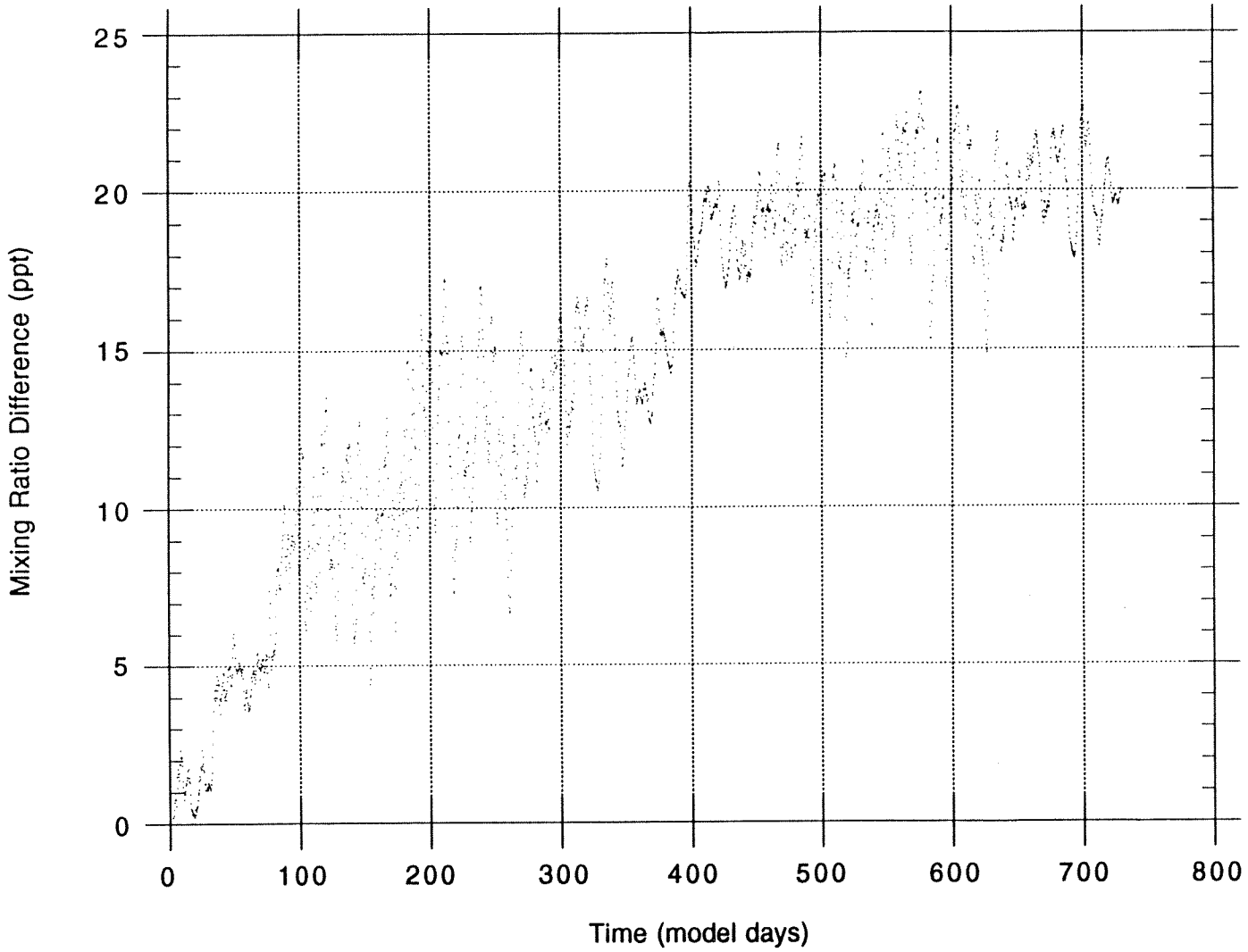


Figure 3a: CTM Model Tracer Mass as a Function of Removal Rate

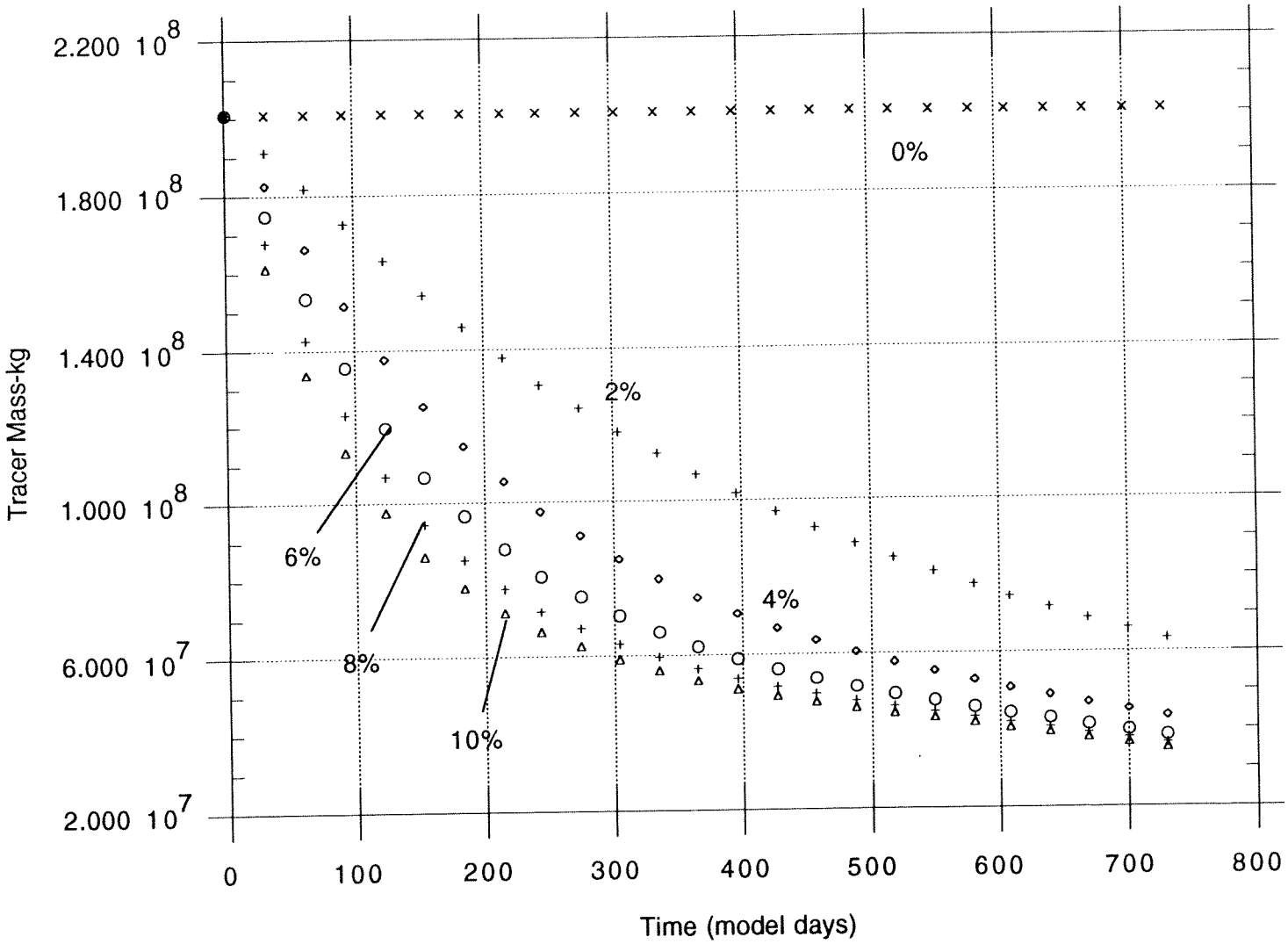


Figure 3b: CTM Model Tracer Mass as Function of Removal Rate

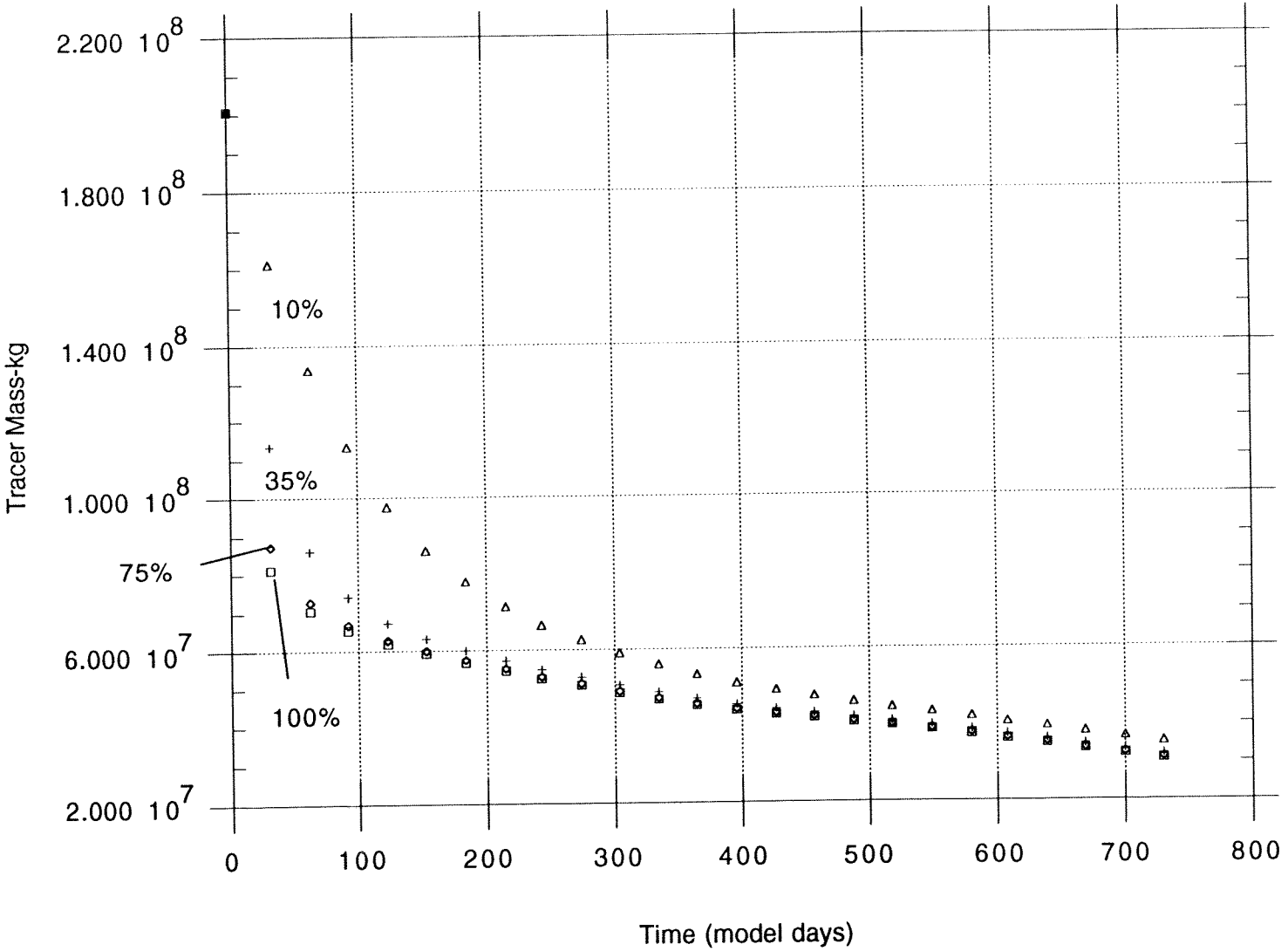


Figure 4a: Instantaneous Lifetime as a Function of Removal Rate

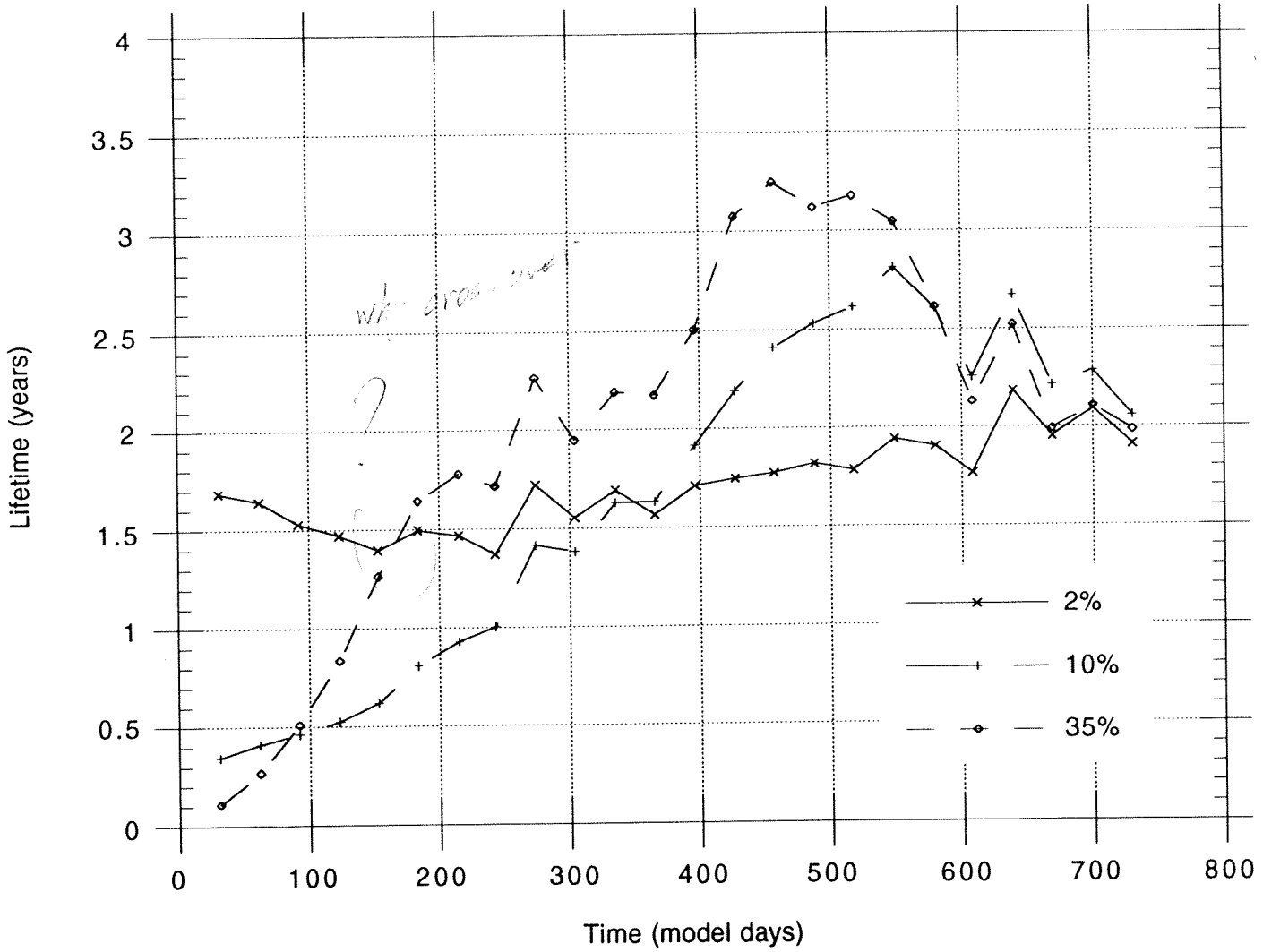


Figure 4b: Instantaneous Lifetime as a Function of Removal Rate

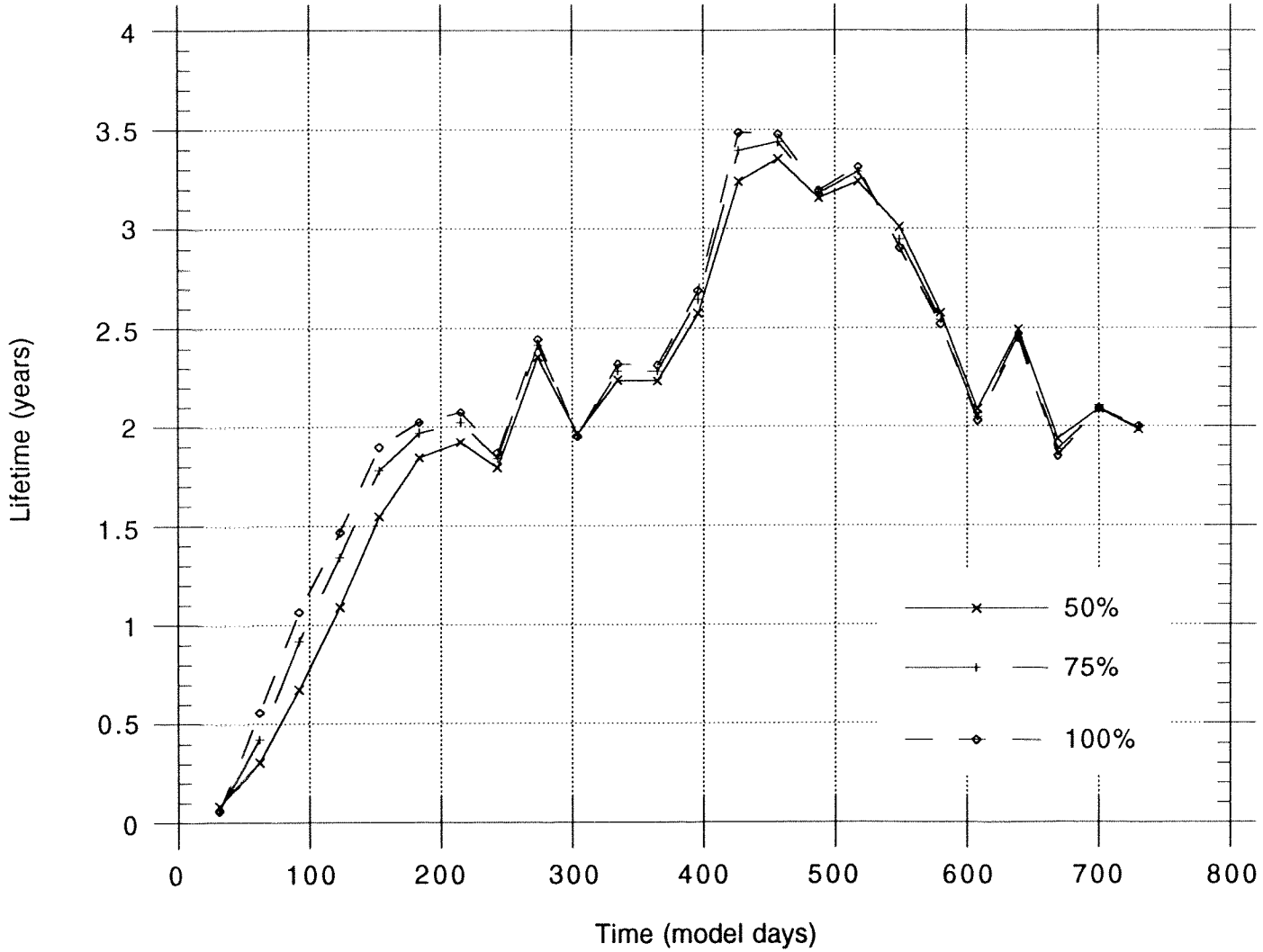


Figure 5a: Time Series of Chlorine Fraction for Layer 1 Emission

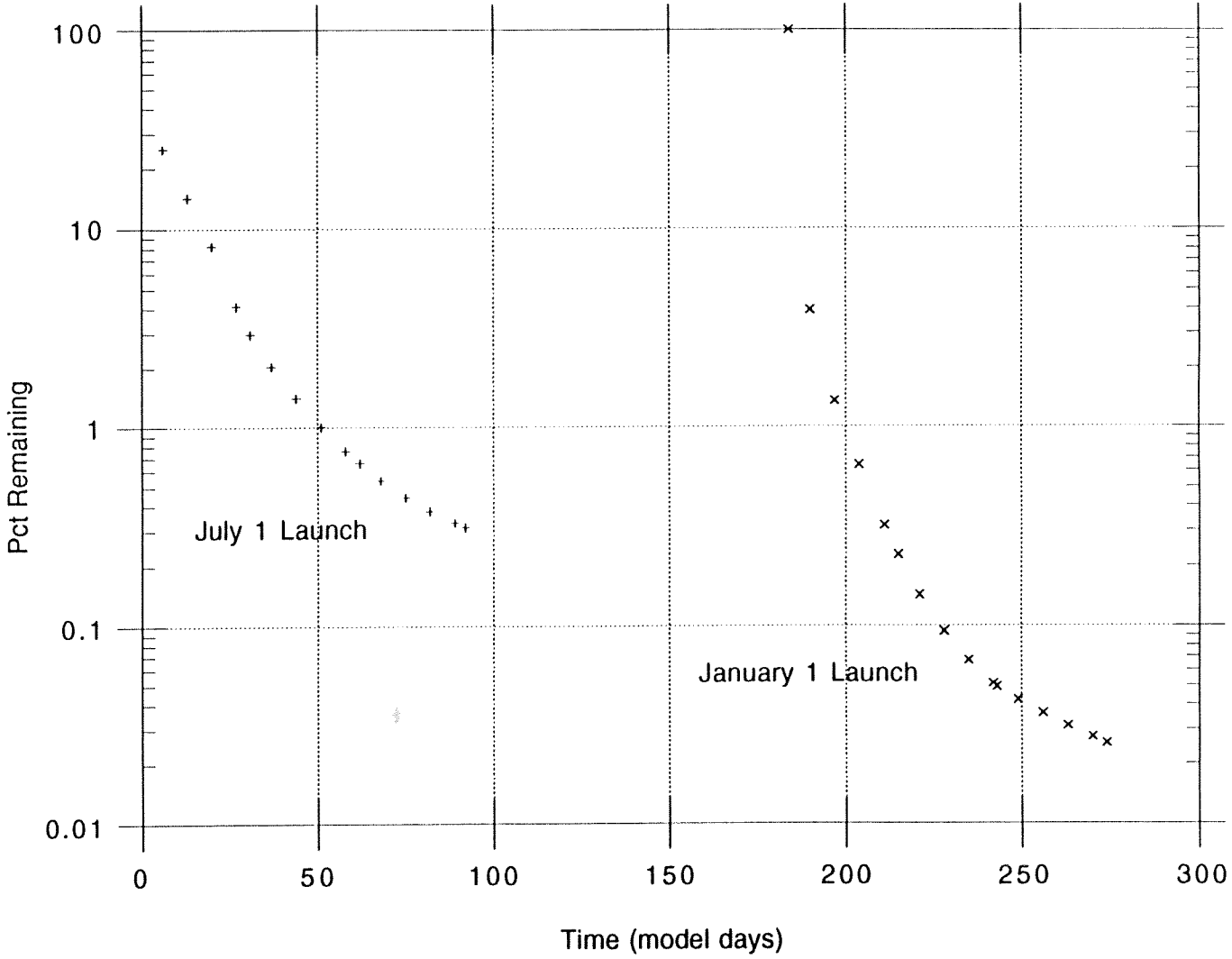


Figure 5b: Time Series of Chlorine Fraction for Layer 4 Emission

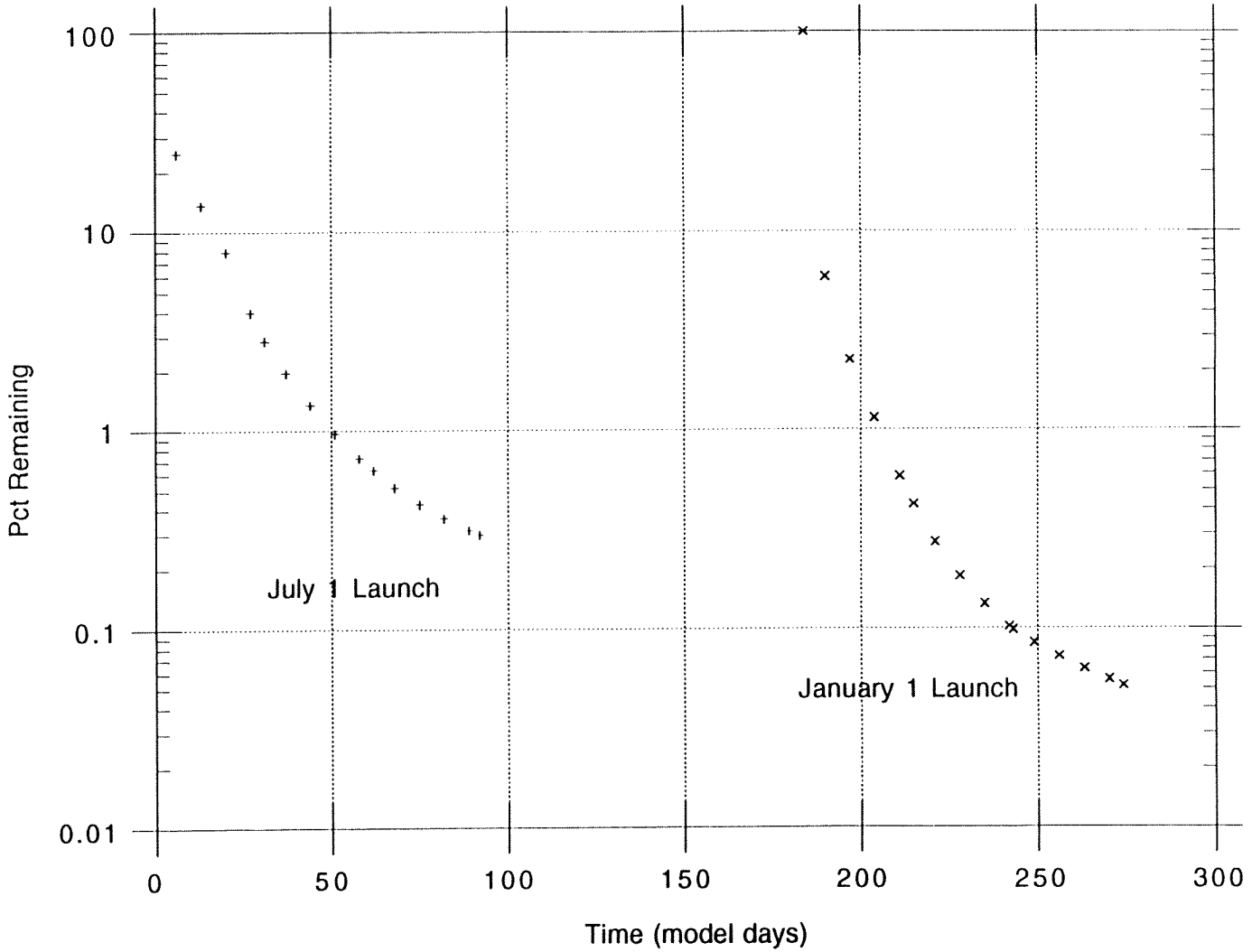
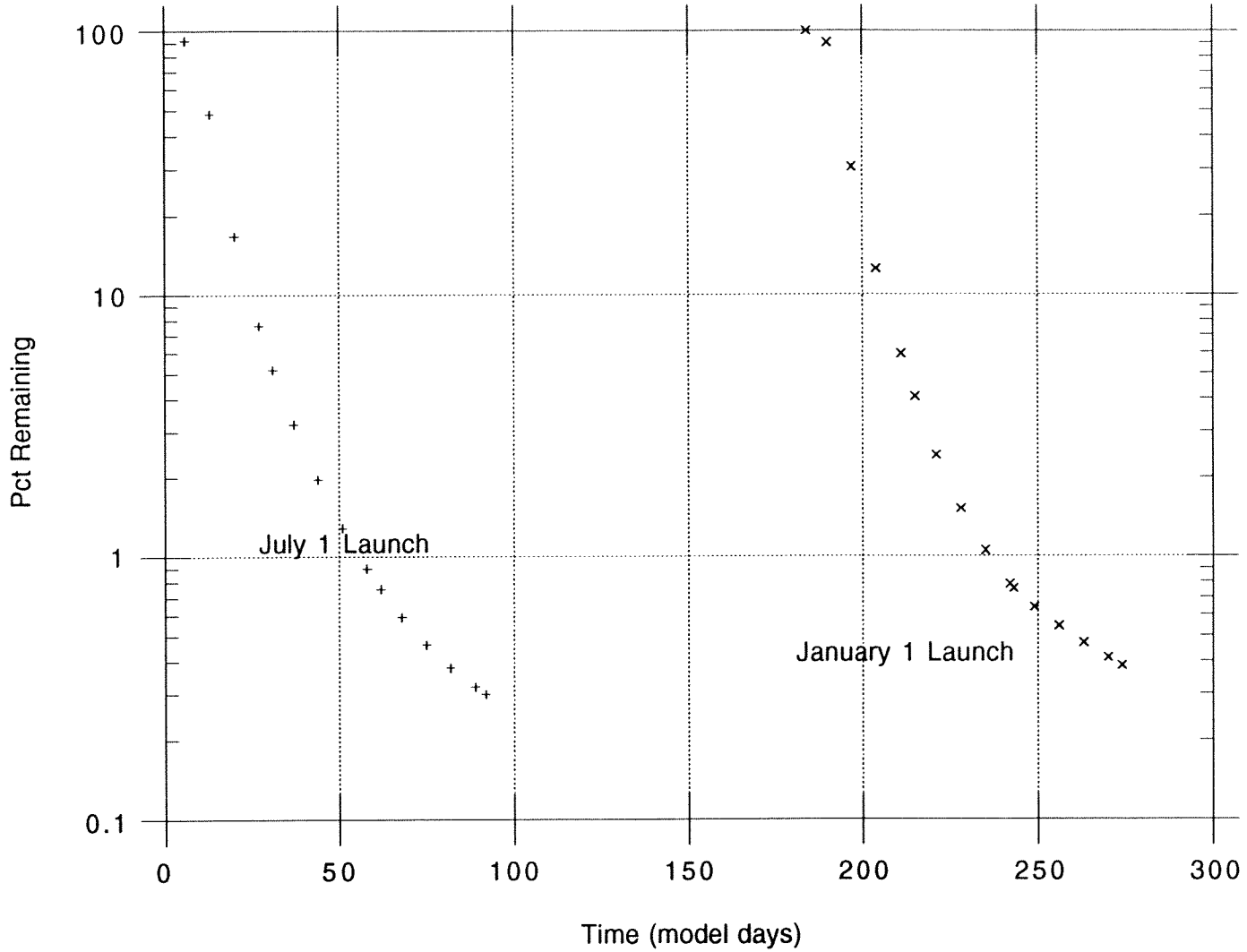


Figure 5c: Time Series of Chlorine Fraction for Layer 7 Emission



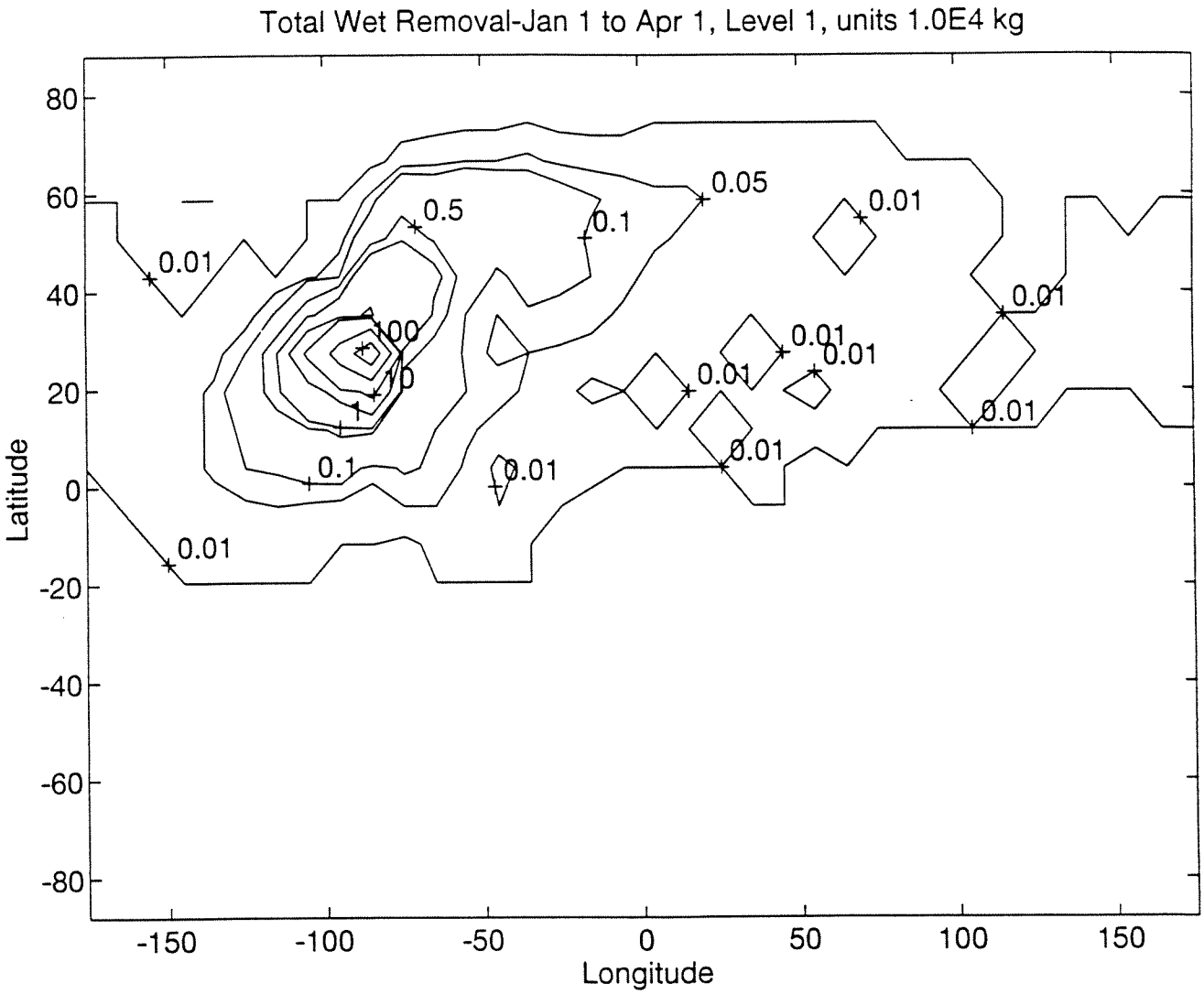


Figure 6: Spatial distribution of surface wet deposition (in kg HCl/grid area) for the three-month simulations with single layer emissions, January launch

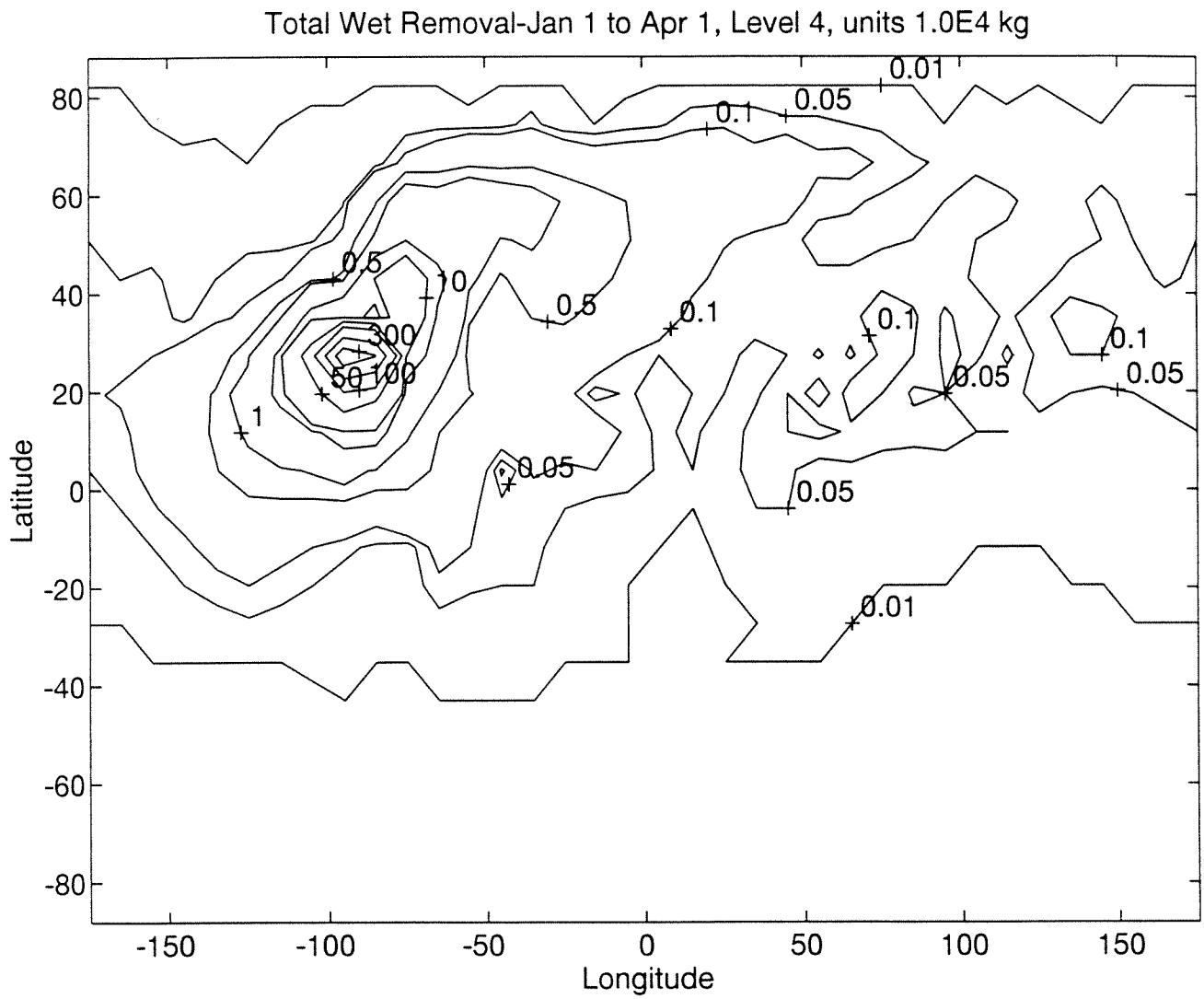


Figure 6: Spatial distribution of surface wet deposition (in kg HCl/grid area) for the three-month simulations with single layer emissions, January launch

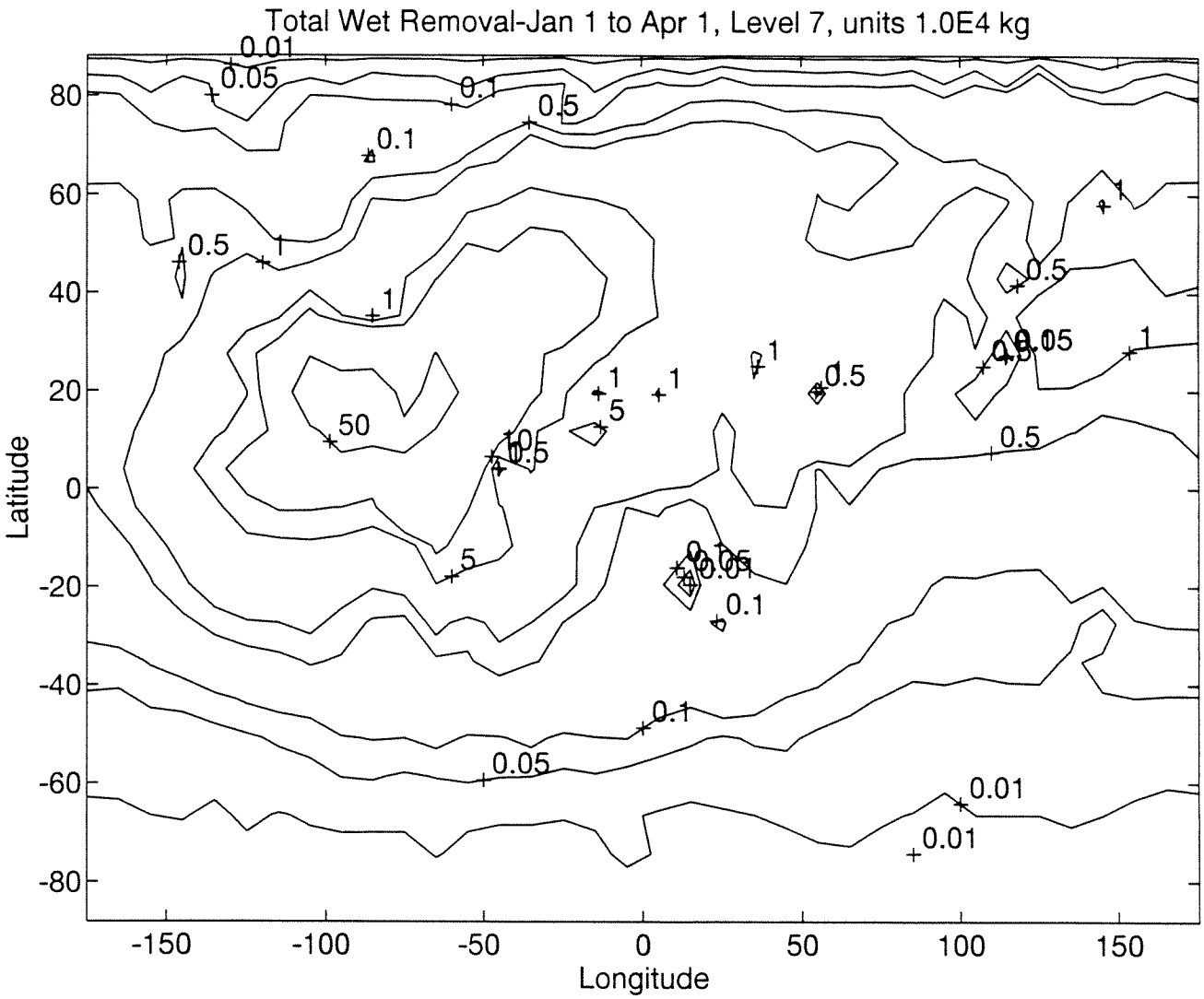


Figure 6: Spatial distribution of surface wet deposition (in kg HCl/grid area) for the three-month simulations with single layer emissions, January launch

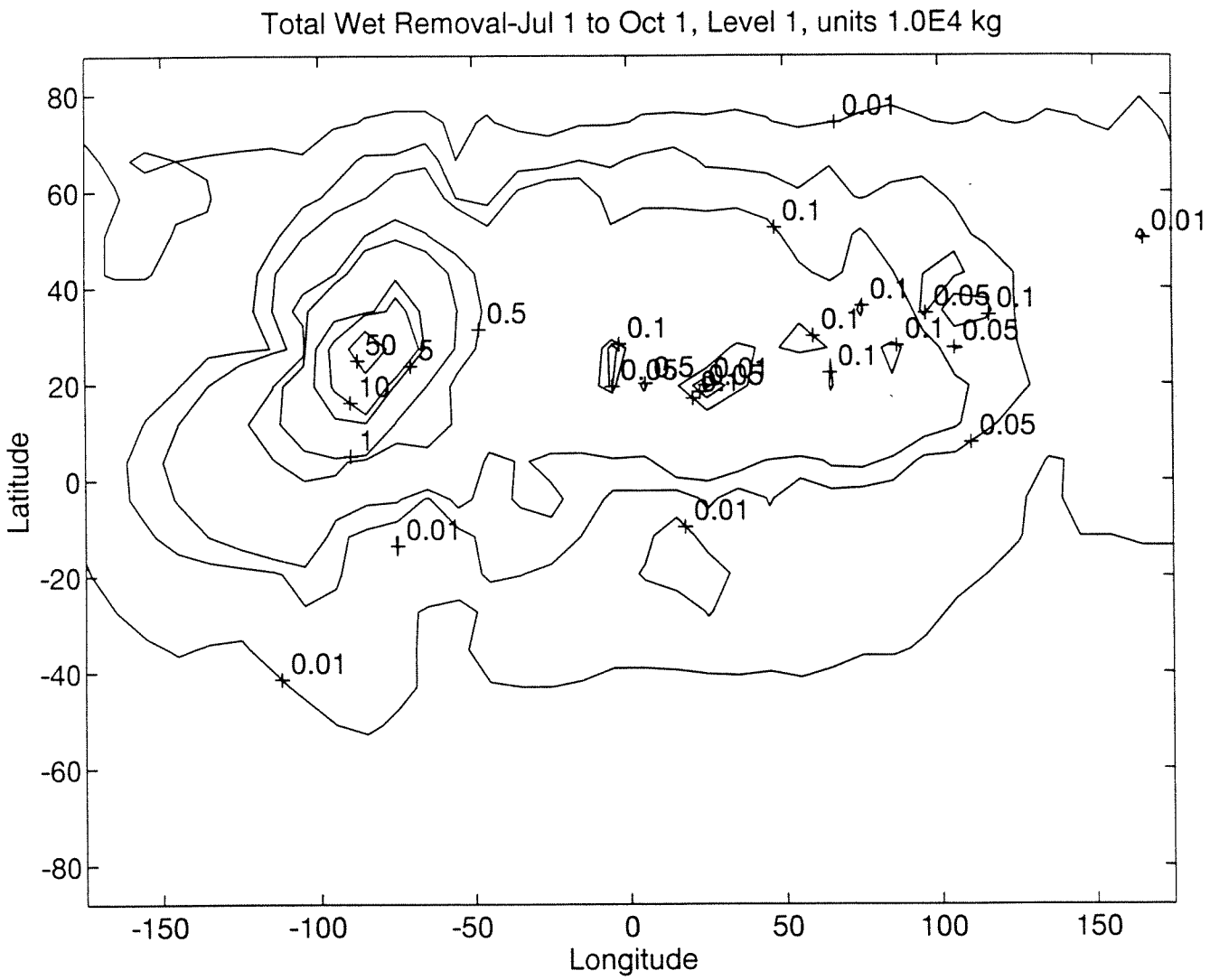


Figure 7: As for Figure 6, July launch

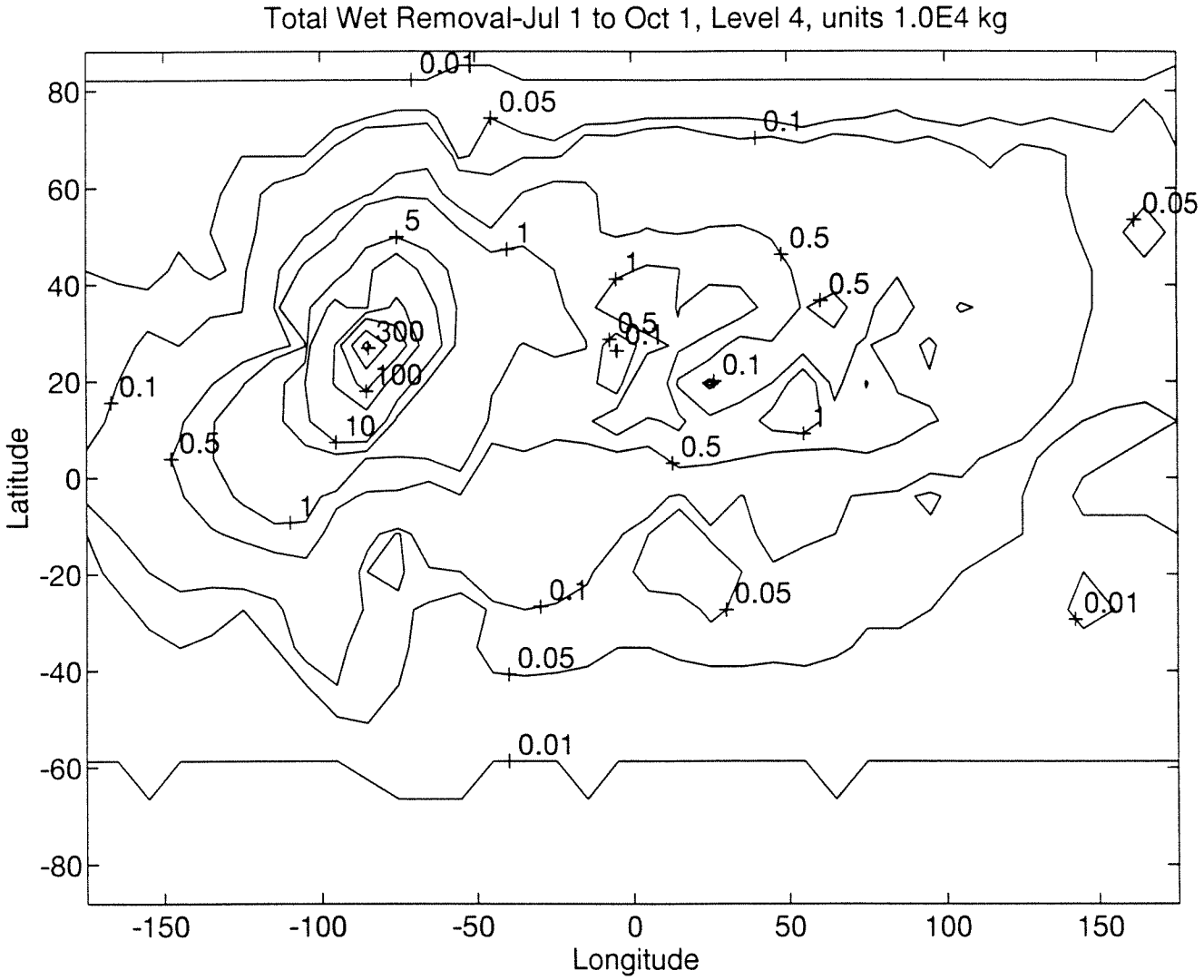


Figure 7: As for Figure 6, July launch

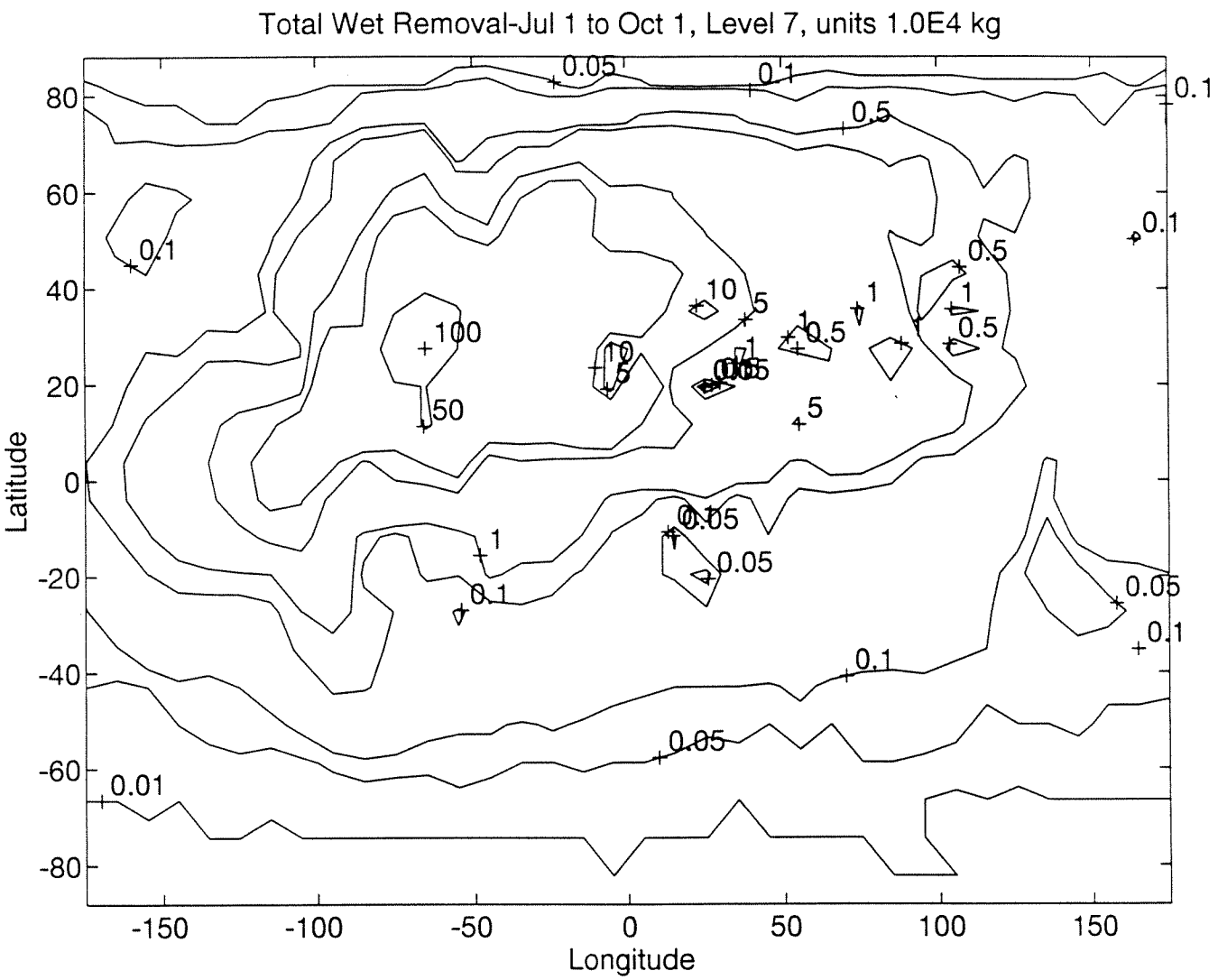


Figure 7: As for Figure 6, July launch

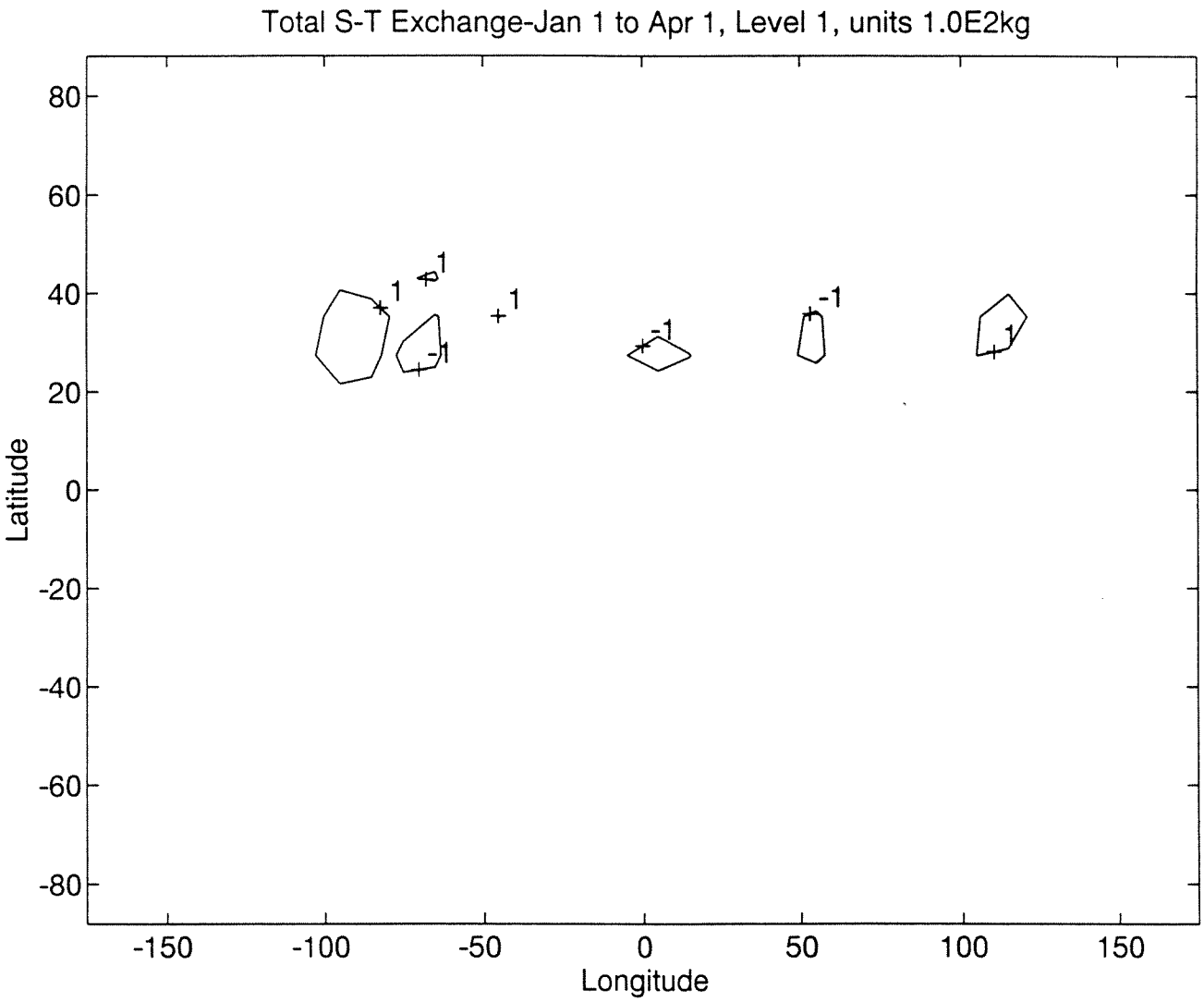


Figure 8: Spatial distribution of transport across diagnostic tropopause (kg HCl/ grid area) for the three-month simulations, with single layer emissions, January launch

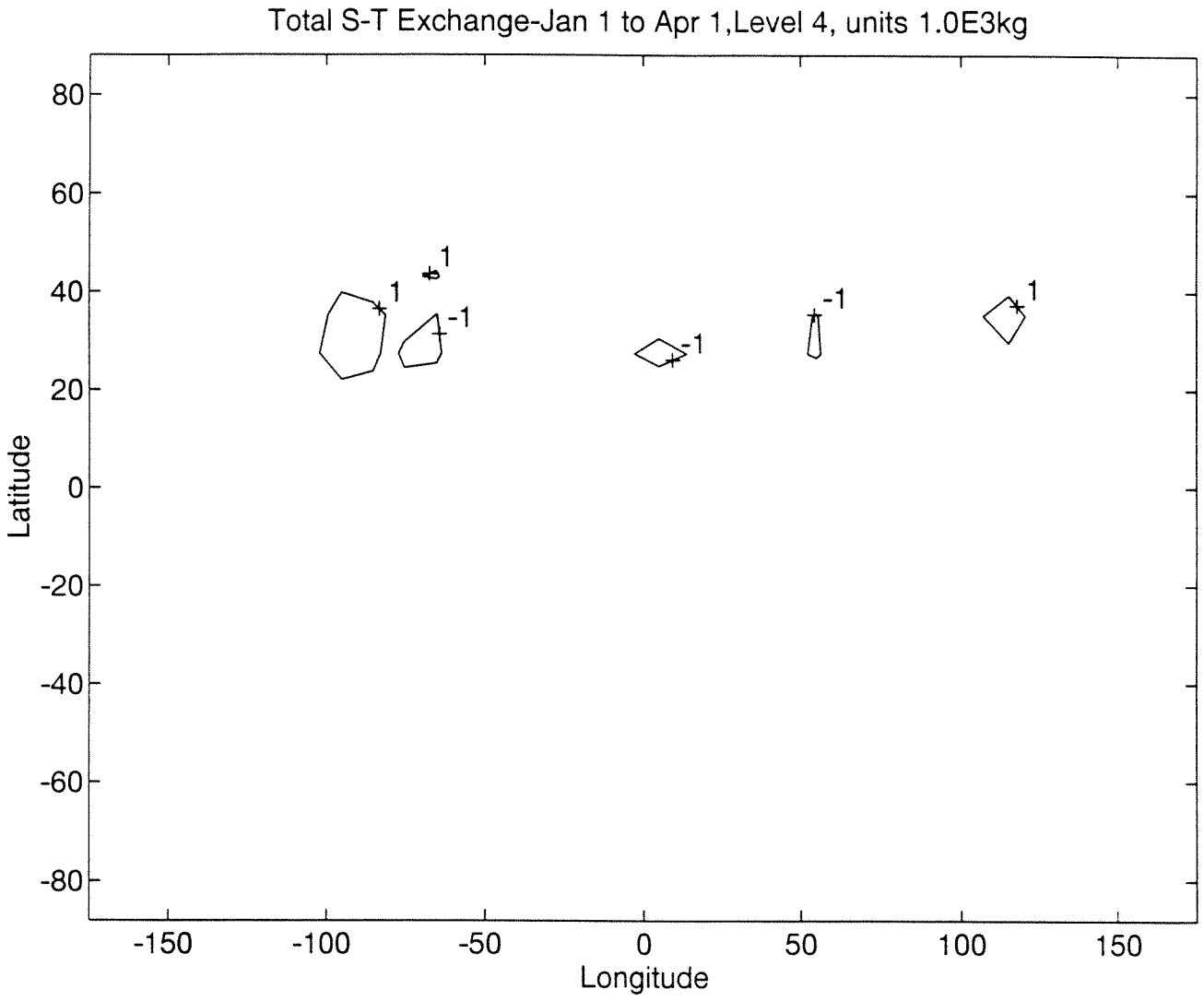
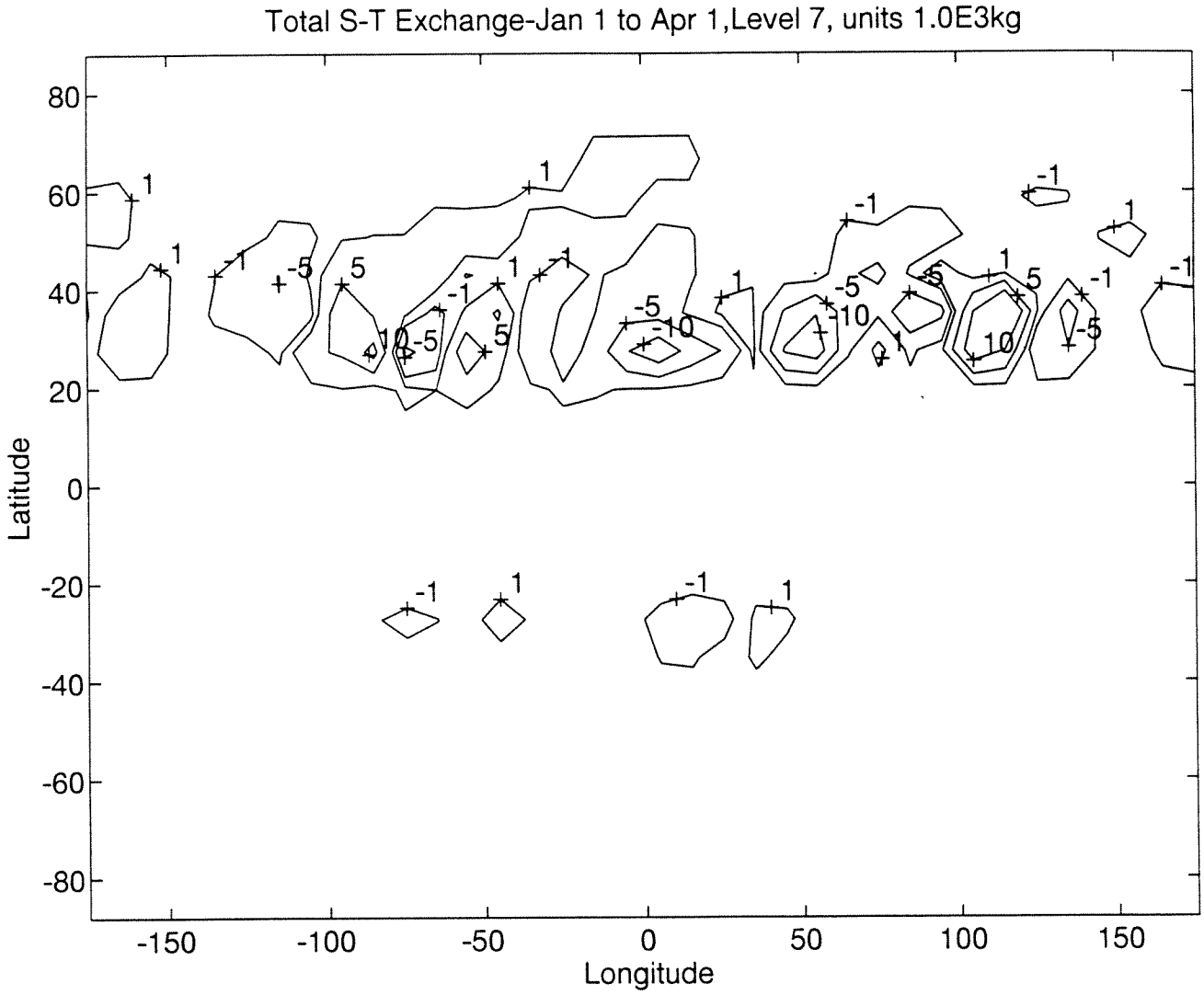


Figure 8: Spatial distribution of transport across diagnostic tropopause (kg HCl/ grid area) for the three-month simulations, with single layer emissions, January launch



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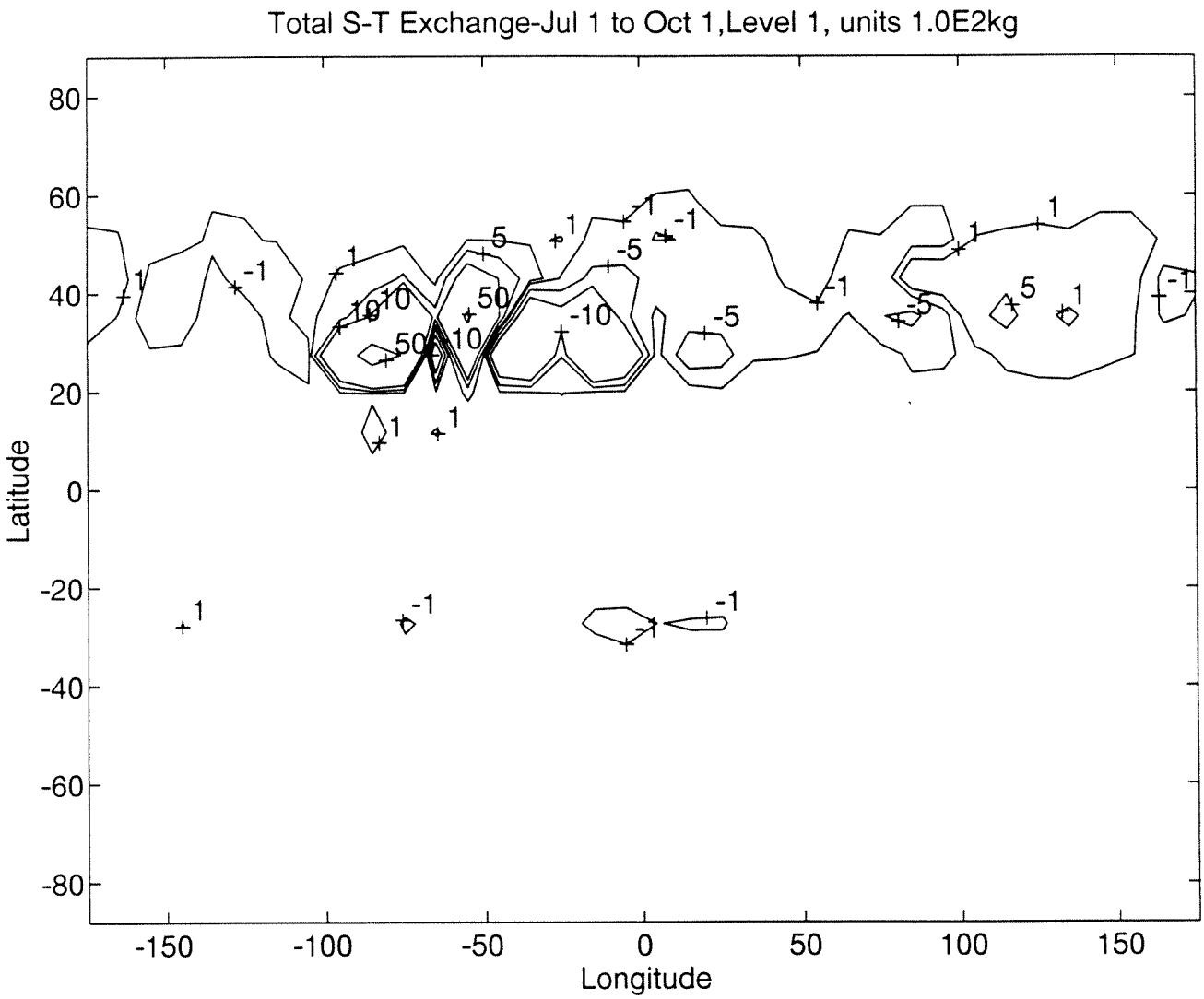

Figure 9: As for Figure 8, July launch

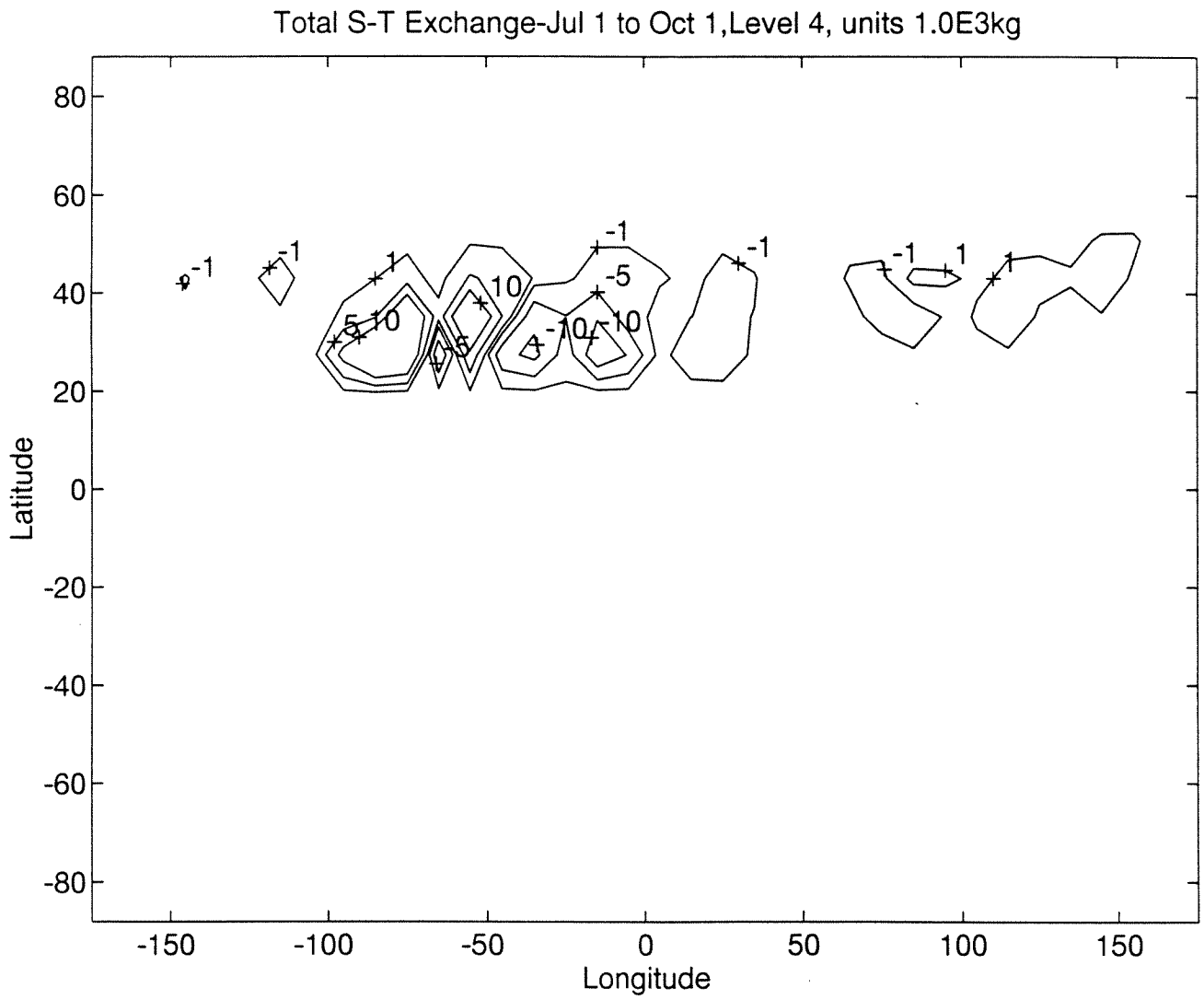

Figure 9: As for Figure 8, July launch

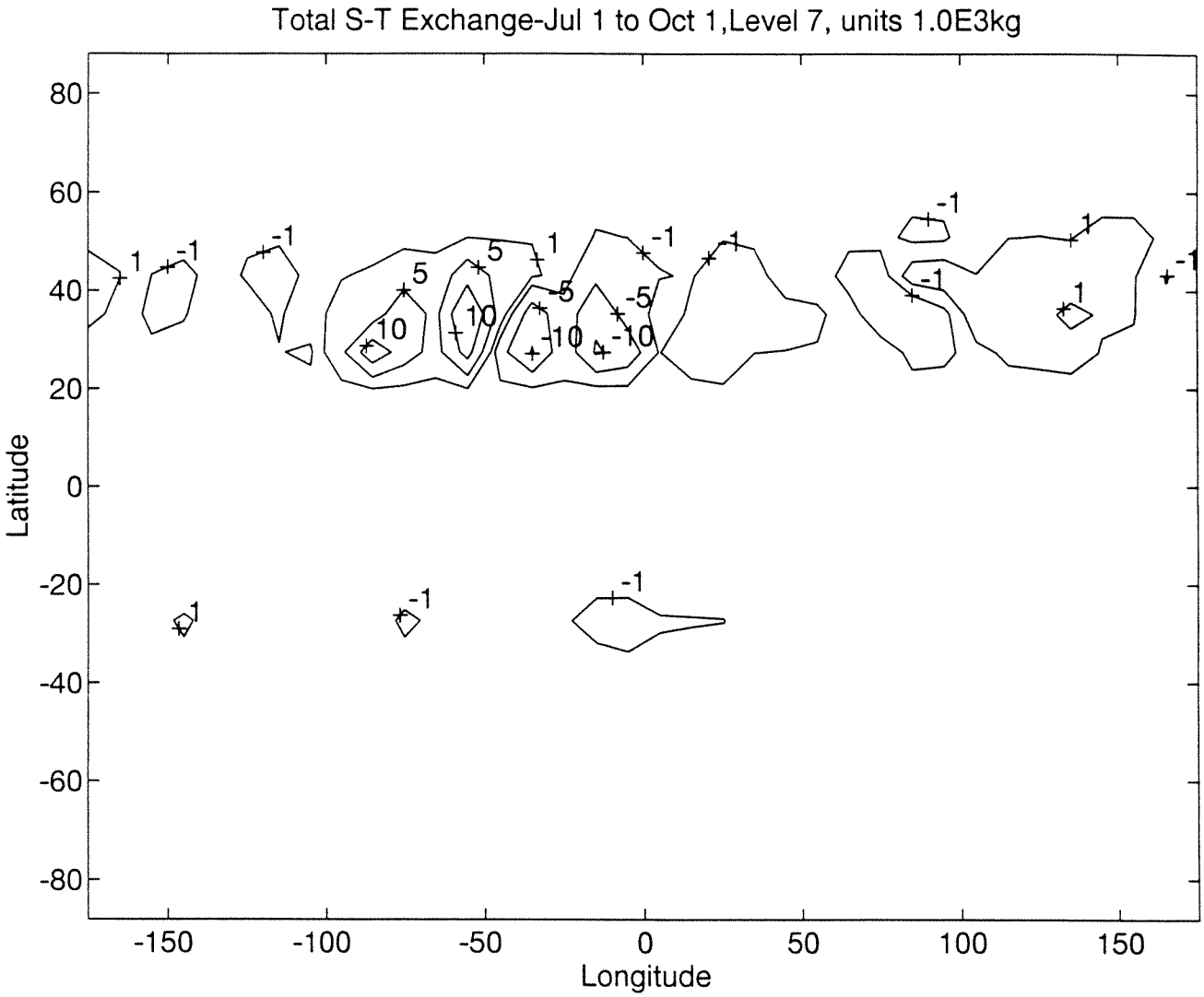

Figure 9: As for Figure 8, July launch